Time-Dependent Electrical Resistance of Transmutational Material With ^{57}Co

Norimasa Yoshimizu

***Abstract*— Transmutational material is studied as a changing electrical resistance over time. A process compatible to complementary metal oxide semiconductors is developed to deposit transmutational material. The material contains the radioisotope ^{57}Co which decays and causes an elemental change, in turn causing a change in electrical resistance over time. Significant increases– nearly a factor of four– in sample resistance over time are observed. Scaling is presented to show that samples that are less than 10 % of typical background exposure could be fabricated on the scale of microelectronics, avoiding detection. Finally, an application is demonstrated of a Wheatstone bridge containing a transmutational sample, leading to timed disabling of a power regulator.**

***Index Terms*— Radioactive materials, time measurement, transmutation.**

I. Introduction

THE use of radioactive materials in technological applications is typically designed to exploit their energetic emissions, such as due to their alpha or beta particles, or gamma rays. Nuclear batteries utilize the heat generated by the radioactive decay and convert it into electrical energy or, more directly, collect charged particles onto capacitive plates [1], [2] or generate electron-hole pairs in semiconducting materials [3]. For time measurement, a radioactive counting clock was reported which counted the beta radiation emitted by ^{63}Ni on an avalanche photodetector [4]. The use of radioisotopes in time measurement is predominated by radiocarbon dating [5]. Radiocarbon dating takes advantage of the decay of ^{14}C into ^{14}N where the relative presence of radioactive ^{14}C is used to determine the age of a sample.

Radiocarbon dating in particular takes advantage of an important distinction in the radioactive decay process. A radioisotope that undergoes decay experiences two distinct effects. First, alpha, beta, or gamma radiation is emitted, helping stabilize the atom. This is the effect used in most of the technologies described above. Second, the nucleus itself usually undergoes change in its atomic element, also called nuclear transmutation. In radiocarbon dating, the radioactive carbon atoms transmutate into nitrogen atoms and thus occur in lower concentrations as time passes. This transmutational effect is used less often in radioisotope applications but offers a unique capability. This paper reports on a study of transmutational material over time and its possible applications. It will show that transmutational material exhibits time-dependent resistivity that can be used for autonomous time measurement, specifically demonstrating a Wheatstone bridge circuit that triggers a timed disabling of a power regulator.

As a radioactive material transmutates over time, the changing elemental composition will cause related changes in bulk properties such as electrical resistance. There have been previous investigations of changes in electrical resistivities in radioactive materials. King et al. studied the changes in resistivity due to self-damage of plutonium, neptunium, and uranium showing resistivity changes of up to 3.5× at 4.5 K [6]. Johnson et al. showed time-dependent changes in resistivity of neutron-irradiated CdS [7], [8]. Müller et al. showed up to 7× net increase in electrical resistivity of ^{241}Am over time [9]. Finally, Rohrlack et al. implanted the radioisotopes ^{67}Ga and ^{71}As into GaAs, generating acceptor and donor sites and observing up to 2.5× change in sheet resistivity [10]. The application of neutron irradiation and subsequent generation of transmutational material is similar to neutron transmutation doping, a technique where dopants can be introduced into semiconducting material more uniformly than thermal or ion beam techniques [11]. These previous studies, except the latter, involved techniques not chosen for making transmutational materials in technological applications. For example, designing an electronic system then irradiating it to generate radioactive material would also generate excess radioactivity, such as within the substrate and surrounding areas. Subsequently etching a thin film as part of a microfabrication process would generate unused radioactive waste material as well, increasing cost and fabrication complexity.

II. Sample Preparation

This paper investigates transmutational material deposited directly onto a substrate by plating methods. Since the material is plated, the radioactive material can be selectively deposited. This isolates the radioactive material to where it is desired and increases the concentration that can be achieved, reducing the total activity that needs to be used. The process developed in this paper is also compatible to complementary metal oxide semiconductors (CMOS), requiring low temperatures. In particular, we study the deposition of ^{57}Co, a radioisotope

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which decays into Fe with a half-life of 272 days. The decay scheme of ^{57}Co proceeds by electron capture, resulting in the excited $J = 5/2^-$ state of Fe at 136 keV above the ground state and typically (88 % probability) emitting 122 keV photons and some subsequent, low energy electrons [12]. Cobalt 57 is commercially available at high specific activity, even at its maximum specific activity of 8500 Ci/g at the time of lot production. These characteristics make ^{57}Co a suitable, practical candidate for producing transmutational material that has the necessary change in resistance with minimal activity and detectability at low cost.

Samples were prepared using high specific activity of ^{57}Co using an electroless plating process developed based on earlier work [13], [14]. The radioactive solutions used were approximately 72 % of maximum specific activity (6,120 Ci/g). The electroless plating solution with radioactive ^{57}Co was a 0.1 M HCl solution containing $CoCl_2$. The pH of the solution was adjusted by adding NaOH then evaporating off some water, resulting in a pH of 1.1. The substrate was bulk 1 cm × 2 cm aluminum masked with 2.5 mil-thick Kapton tape with a 0.5 mm ×0.5 mm opening for cobalt deposition and a 0.75 mm ×0.75 mm opening for substrate contact. The substrate was maintained at 120 °C. A micropipette was used to deposit 1 μL of solution at a time. The solution, after each deposit, leaves behind visible, solid crystals of NaCl which are removed in water. In total, 1 mCi are deposited per sample.

The plating of cobalt onto aluminum was confirmed by studying the equivalent, non-radioactive deposition process. At each step during drop-by-drop deposition, the solution dries and leaves behind solid NaOH. When the cobalt is depositing onto the aluminum substrate, the NaOH dries white in color. However, when cobalt is not depositing onto the aluminum and remains in the solution, the NaOH will dry with the light blue color characteristic of anhydrous $CoCl_2$. In addition, comparing deposition with and without $CoCl_2$ shows obvious, visual differences on the substrate. Deposition with $CoCl_2$ results in a dark substrate surface whereas deposition without it retains a shiny, metallic appearance. Finally, energy dispersive X-ray spectroscopy measurements on a Tescan Mira3 field emission scanning electron microscope show the presence of significant amounts of cobalt on the substrate.

Table I shows the ratio of atomic concentrations of cobalt to aluminum and the same ratio per μL of deposition volume. There is a significant increase in the concentration of cobalt relative to aluminum, but it remains fairly constant per volume of solution. The deposition of radioactive cobalt was also confirmed by the radioactivity measured on the samples using a gamma detector. Control samples using the non-radioactive deposition process were also used to confirm that no changes in resistance are seen over time.

TABLE I
ELEMENTAL CONCENTRATIONS OF COBALT DEPOSITION SAMPLES

Deposition solution volume (μL)	Ratio of atomic concentrations, Co/Al	Ratio of atomic concentrations, Co/Al, per volume of solution
3	9.2 %	3.1 %/μL
6	15.2 %	2.5 %/μL
9	42.3 %	4.7 %/μL
12	43.9 %	3.7 %/μL
15	95.3 %	6.4 %/μL
18	99.1 %	5.5 %/μL

Atomic concentrations of cobalt on 0.5 × 0.5 mm^2 area of aluminum substrate after deposition, as measured by energy dispersive X-ray spectroscopy.

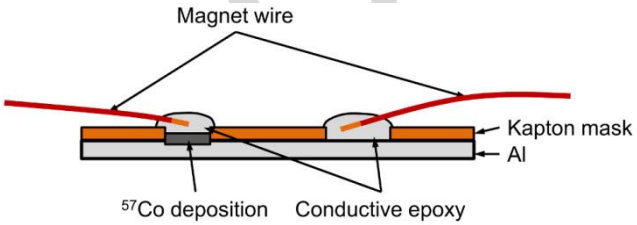

Fig. 1. Setup for resistance measurement of transmutational material. A Kapton tape mask was used to mask the deposition of ^{57}Co onto an Al substrate. Conductive silver epoxy was used to connect magnet wires to the substrate.

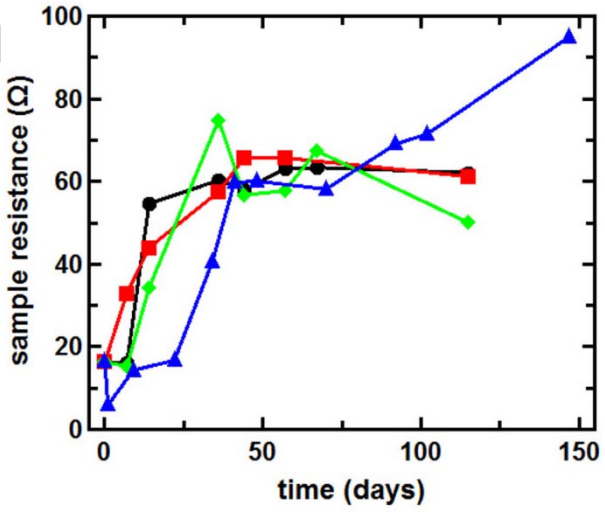

Fig. 2. Time-dependent resistance of four samples, showing data after series resistance is removed then scaled so that initial resistances are equal to the unadjusted curve depicted with red squares.

III. SAMPLE MEASUREMENT

The transmutational samples were measured directly through wires that were attached by conductive epoxy onto the ^{57}Co deposition area and the substrate; see Fig. 1. A source meter was used to perform a four-point measurement of the resistance through the samples at 10 mA of current. The measurements of the transmutational samples are shown in Fig. 2.

The raw data used for Fig. 2 showed variation among the four samples. Note that the blue curve has different sampling days as it was tested in a separate fabrication and test run. The initial resistances of the samples varied from 7.4 Ω to 21.8 Ω due to two main causes. First, there is variation in the series resistance to the sample incurred by the substrate, silver epoxy, and magnetic wires. Second, there is variation caused by the

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deposition process. Only about 160 ng of ^{57}Co are deposited per sample along with 62 ng of nonradioactive isotopes of Co. The film is not contiguous and its effect on total sample resistance will vary based on its concentration across the sample. For example, a deposition whose coverage is sparse or thin will contribute smaller changes in resistance, whereas deposition that is both even and thicker will contribute larger changes. Therefore, the total sample resistance of a sample is assumed to be,

$$R_{\text{total}}(t) = R_0 + \alpha R_{\text{trans}}(t) \quad (1)$$

where R_0 is the series resistance due to the substrate, silver epoxy, and magnet wires, and α scales the resistance R_{trans} (t) of the transmutational film due to deposition process variations.

Fig. 2 shows the data after removing these variations. The data depicted in red squares is unchanged. The other data curves were generated by removing an offset, thereby removing the constant R_0 in Eq. 1, then scaling with a multiplicative constant so that the initial resistances all match at 16.3 Ω. The offset parameters are adjusted so that each data curve best matches the data depicted by the red squares by minimizing the mean squared error. The resulting curves show significant increase in resistance by a factor of 3.8 ± 0.4 after only 42% of half-life has passed.

IV. Discussion

The data show that large changes in resistance can be generated in transmutational samples, implying that very small radioactivity could be used in many applications. Doing so reduces the risk to human and environmental health, cost, and detectability of the radioactive material. In a clandestine application, it is desirable to prevent the detection of transmutational material using radiation detectors. Therefore, we consider as a stringent limit on radioactivity a level which would be difficult to distinguish from background. For ^{57}Co, a gamma emitter, typical background for a gamma detector is 10 μR/hr. We take as a limit of detectability ten percent of background, or 1 μR/hr. We assume that the exposure from a ^{57}Co source is primarily due to its 122 keV gamma rays and use typical physical parameters: ion generation in air at 33.8 eV per pair, X-ray mass attenuation coefficient in dry air at 122 keV of 2.4×10^{-2} cm^2/g, a detector with a 1 cm radius, and a distance of 2.54 cm between the radioactivity and the detector. As a result, 11 nCi of ^{57}Co generates an exposure of 1 μR/hr at a distance of 2.54 cm. Using the gamma ray dose constant of 0.151 rem/hr/Ci at one meter yields a dose of 2.6 μ rem/hr at 2.45 cm and, for comparison, the U.S. Nuclear Regulatory Commission exempt quantity for ^{57}Co is 100 μ Ci, or about 10,000 times more activity than a single, 11 nCi transmutational device [15].

Fig. 3 shows the scaling of device geometries by reducing device radioactivity using ^{57}Co. The range of areas and thickness required for various radioactivities and specific activities are shown. Note that these dimensions are compatible with typical microfabrication parameters. The activities could be further decreased by reducing the specific activity and thus

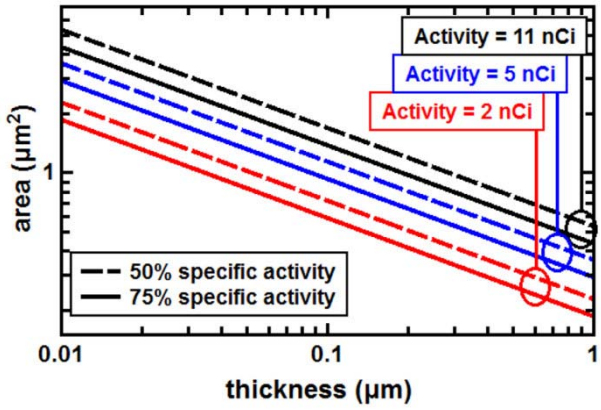

Fig. 3. Parameters for fabricating low activity devices using 50 or 75% specific activity ^{57}Co with dimensions of micrometers in width and hundreds of nanometers in thickness. Devices with activities of 2, 5, or 11 nCi are shown, where the latter generates about 1 μR/hr at 2.54 cm for a typical gamma detector, or about 10% of background.

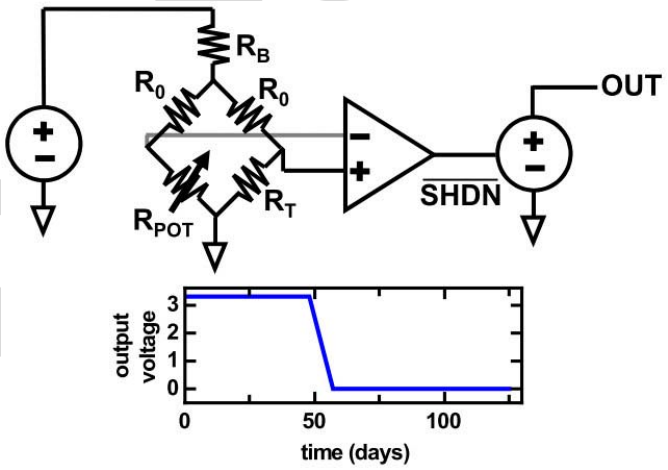

Fig. 4. Schematic of example application of transmutational material, where a Wheatstone bridge triggers a signal to shut down a power regulator. Data below shows output voltage from printed circuit board prototype over time.

reducing the net change in resistance as a tradeoff, as the data show that even small amounts of transmutation can cause significant changes in resistance. In summary, it may be feasible to scale down the fabrication processes discussed in this paper to create micron and submicron-scale devices which are radiologically safe and very difficult to detect.

The use of transmutational properties may have advantages over other technologies where radioactive materials act as a timing mechanism. Measuring radioactive decay requires extra circuitry and significant activity to generate sufficient signal strength and particle counts to reach the required timing accuracy. For comparison, a 1 mCi source of ^{57}Co only generates 720 nW of energy. However, if a transmutational material can increase the series impedance of a device consuming 20 mA at 3.3 V by 20 Ω (12 %), the power delivered to the device would be reduced by 14 mW (21 %). Such changes can also be measured more easily and precisely by comparators or other differential circuits.

Applications which may benefit would be those that require an independent, self-powered, and time-dependent process.

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Note that the transmutation process, due to its underlying physics, is largely unaffected by external electromagnetic, mechanical, or thermal forces and thus reliable and robust against many physical effects. In a simple application the resistance across a transmutational device could be used as a measurement of time. That measurement could be recorded or, if integrated into a circuit, used to cause failure or malfunction in an electronic device. Note that hardware Trojan taxonomies do not yet seem to include a classification for purely time-dependent attacks [16]. Fig. 4 shows such an example where a transmutational resistor was used to cause a power regulator to shut down after some time had passed. A Wheatstone bridge has a bias resistor R_B, constant leg resistances R_0, a transmutational resistor R_T, and an adjustable resistance R_{POT} which is set relative to R_T to define a time until shutdown of the regulator. The data at the bottom of Fig. 4 is empirical data using a transmutational sample to cause a commercial 3.3 V power regulator to shut down after seven weeks.

V. Conclusion

This paper demonstrated a time-dependent resistor using transmutational material with a 3.8× increase in resistance. A CMOS-compatible, selective deposition process was developed. Scaling was studied to show compatibility with microelectronic geometries. Finally, an application involving a Wheatstone bridge used to trigger shutdown in a power regulator was demonstrated.

Acknowledgment

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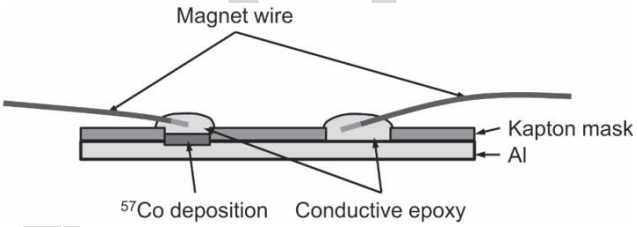

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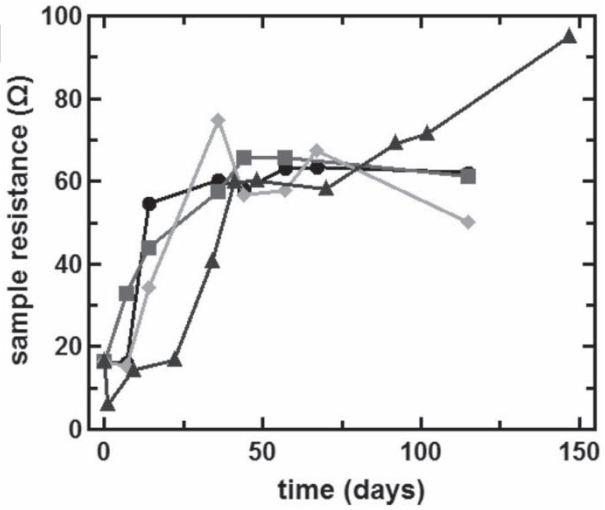

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Fig. 2 shows the data after removing these variations. The data depicted in red squares is unchanged. The other data curves were generated by removing an offset, thereby removing the constant R_0 in Eq. 1, then scaling with a multiplicative constant so that the initial resistances all match at 16.3 Ω. The offset parameters are adjusted so that each data curve best matches the data depicted by the red squares by minimizing the mean squared error. The resulting curves show significant increase in resistance by a factor of 3.8 ± 0.4 after only 42% of half-life has passed.

IV. Discussion

The data show that large changes in resistance can be generated in transmutational samples, implying that very small radioactivity could be used in many applications. Doing so reduces the risk to human and environmental health, cost, and detectability of the radioactive material. In a clandestine application, it is desirable to prevent the detection of transmutational material using radiation detectors. Therefore, we consider as a stringent limit on radioactivity a level which would be difficult to distinguish from background. For ^{57}Co, a gamma emitter, typical background for a gamma detector is 10 μR/hr. We take as a limit of detectability ten percent of background, or 1 μR/hr. We assume that the exposure from a ^{57}Co source is primarily due to its 122 keV gamma rays and use typical physical parameters: ion generation in air at 33.8 eV per pair, X-ray mass attenuation coefficient in dry air at 122 keV of 2.4×10^{-2} cm^2/g, a detector with a 1 cm radius, and a distance of 2.54 cm between the radioactivity and the detector. As a result, 11 nCi of ^{57}Co generates an exposure of 1 μR/hr at a distance of 2.54 cm. Using the gamma ray dose constant of 0.151 rem/hr/Ci at one meter yields a dose of 2.6 μ rem/hr at 2.45 cm and, for comparison, the U.S. Nuclear Regulatory Commission exempt quantity for ^{57}Co is 100 μ Ci, or about 10,000 times more activity than a single, 11 nCi transmutational device [15].

Fig. 3 shows the scaling of device geometries by reducing device radioactivity using ^{57}Co. The range of areas and thickness required for various radioactivities and specific activities are shown. Note that these dimensions are compatible with typical microfabrication parameters. The activities could be further decreased by reducing the specific activity and thus

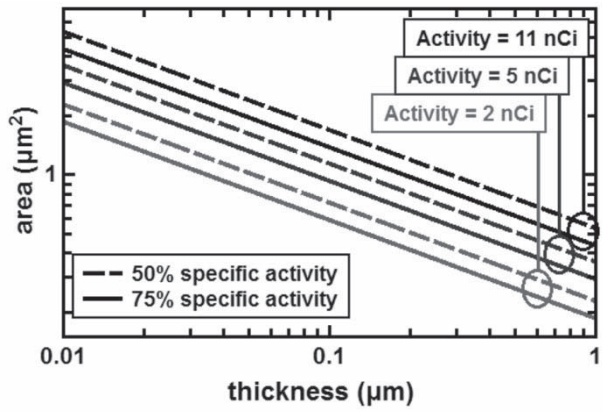

Fig. 3. Parameters for fabricating low activity devices using 50 or 75% specific activity ^{57}Co with dimensions of micrometers in width and hundreds of nanometers in thickness. Devices with activities of 2, 5, or 11 nCi are shown, where the latter generates about 1 μR/hr at 2.54 cm for a typical gamma detector, or about 10% of background.

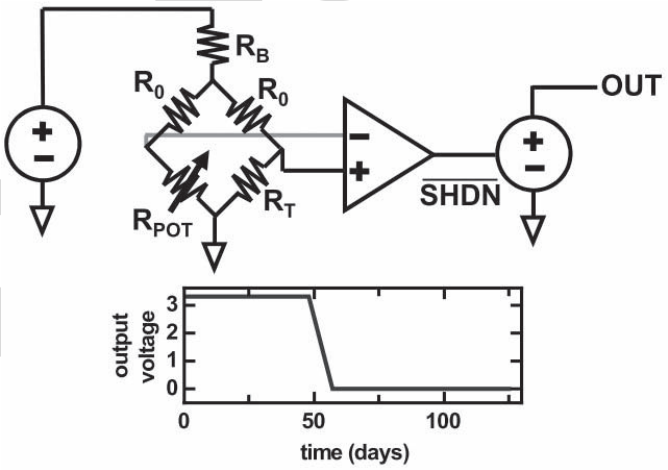

Fig. 4. Schematic of example application of transmutational material, where a Wheatstone bridge triggers a signal to shut down a power regulator. Data below shows output voltage from printed circuit board prototype over time.

reducing the net change in resistance as a tradeoff, as the data show that even small amounts of transmutation can cause significant changes in resistance. In summary, it may be feasible to scale down the fabrication processes discussed in this paper to create micron and submicron-scale devices which are radiologically safe and very difficult to detect.

The use of transmutational properties may have advantages over other technologies where radioactive materials act as a timing mechanism. Measuring radioactive decay requires extra circuitry and significant activity to generate sufficient signal strength and particle counts to reach the required timing accuracy. For comparison, a 1 mCi source of ^{57}Co only generates 720 nW of energy. However, if a transmutational material can increase the series impedance of a device consuming 20 mA at 3.3 V by 20 Ω (12 %), the power delivered to the device would be reduced by 14 mW (21 %). Such changes can also be measured more easily and precisely by comparators or other differential circuits.

Applications which may benefit would be those that require an independent, self-powered, and time-dependent process.

IEEE Proof

Note that the transmutation process, due to its underlying physics, is largely unaffected by external electromagnetic, mechanical, or thermal forces and thus reliable and robust against many physical effects. In a simple application the resistance across a transmutational device could be used as a measurement of time. That measurement could be recorded or, if integrated into a circuit, used to cause failure or malfunction in an electronic device. Note that hardware Trojan taxonomies do not yet seem to include a classification for purely time-dependent attacks [16]. Fig. 4 shows such an example where a transmutational resistor was used to cause a power regulator to shut down after some time had passed. A Wheatstone bridge has a bias resistor R_B, constant leg resistances R_0, a transmutational resistor R_T, and an adjustable resistance R_{POT} which is set relative to R_T to define a time until shutdown of the regulator. The data at the bottom of Fig. 4 is empirical data using a transmutational sample to cause a commercial 3.3 V power regulator to shut down after seven weeks.

V. Conclusion

This paper demonstrated a time-dependent resistor using transmutational material with a 3.8× increase in resistance. A CMOS-compatible, selective deposition process was developed. Scaling was studied to show compatibility with microelectronic geometries. Finally, an application involving a Wheatstone bridge used to trigger shutdown in a power regulator was demonstrated.

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